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ULTRAFAST SPECTROSCOPY OF GaAs UNDER MAGNETIC FIELD

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Ultrafast Spectroscopy of GaAs Under Magnetic Field

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Abstract:

Surprising and novel results are obtained for both the linear and the nonlinear optical response of GaAs under magnetic field. Using a variety of spectroscopic techniques, we measure field dependent effects due to Coulomb correlation.

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Ultrafast Spectroscopy of GaAs Under Magnetic Field

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Application of a magnetic field to semiconductors confines and quantizes electronic states with momentum \(k\) perpendicular to \(B\), thereby reducing the dimensionality by two. The 2D to 0D transition has been investigated in 2-D quantum wells under magnetic field. [1,2] The purpose of our work is to study the 3D to 1D transition in a bulk crystal under field. We observe, in high quality GaAs, surprising effects in both linear and transient nonlinear optical response. We attribute these effects to Coulomb correlation, whose importance is well known. [3]

Fig.(1a) illustrates the dramatic effect of Coulomb correlation on the linear absorption of GaAs. [4] For \(B=0\)T the hh and lh excitons are followed by a smooth continuum of states. When \(B\neq0\) the lowest exciton resonances remain almost unchanged, while pronounced asymmetric resonances appear at higher energy.
These are Fano resonances arising from coupling between the discrete magnetoexciton at the edge of the n>1 Landau transition with the energetically degenerate 1D-continua of n'<n Landau levels (LL). Numerical calculations were performed modeling the semiconductor by two parabolic bands in the effective mass approximation. Fig.(1b,c,d) shows results for linear response at B=6T without (b), with intra-LL (c), and with intra- and inter-LL Coulomb interaction (d). It clearly demonstrates that the origin of the Fano interference is a Coulomb correlation induced coupling.

Transient nonlinear optical response was measured via four-wave mixing (FWM) experiments using a 100fs Ti:sapphire laser. Time-integrated (TI), time-resolved (TR) and spectrally resolved measurements of the FWM emission were performed for magnetic fields between B=0-12T. The TI and TR FWM response of the lowest order Lorentzian-magnetoexciton is shown in Fig.(2). With increasing B, the TI-FWM signal clearly develops a strong component at negative time delay, which is a signature of Coulomb correlation. This component disappears for higher excitation density, consistent with increased screening. Further information is obtained from the TR-FWM measurement, which shows a non-exponential temporal evolution with a lengthening decay for increasing B. These results are in striking contrast with those from 2-D systems, where Coulomb correlation is quenched for increasing B.
For the first time, we have measured the transient response of a Fano resonance. We find that it is anomalous, when compared to the well-known Lorentzian resonance response. [5] Fig.(3) summarizes this for B=10T. The TR-FWM measurement decays on a long time-scale ~500fs, consistent with the linewidth and shape of the power spectrum. Very surprisingly, however, the TI-FWM decays instantaneously, as fast as the autocorrelation trace of the laser. Fano resonances are, in fact, structured continua, but this does not suffice to explain their transient nonlinear behavior. Calculations based on an “atomic” Fano model, i.e. with a constant coupling matrix element, indicate that the linear and nonlinear polarizations decay on the same time scale. Again, Coulomb correlation, the cause of the Fano interference in semiconductors, governs their nonlinear response and seems to play a determinant role.

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References:


[3] See, for example, "Optics of Semiconductor Nanostructures," ibid.


Captions:

Figure 1: (a) Measured linear absorption of GaAs at T=1.6K, taken for σ-circularly polarized light. Here strain has lifted the degeneracy in the valence band, so the light hole (lh) and heavy hole (hh) exciton appear split. Theoretical calculations for B = 6T: (b) without; (c) with intra-LL; (d) with intra- and inter-LL Coulomb interaction.

Figure 2: For excitation at the lowest order magnetoexciton: (a) TI-FWM at excitation density N ≈ 10^{16} cm^{-3} and for field strengths B = 10T (top), 6T, 2T, 0T (bottom); (b) TR-FWM at time delay Δt=0, with N ≈ 3x10^{16} cm^{-3}, and for B = 10T (top), 8T, 4T, 0T (bottom).

Figure 3: Transient response of a Fano resonance at B = 10T and excitation density N ≈ 4x10^{15} cm^{-3}: TR-FWM at Δt=0 vs. TI-FWM, with the laser autocorrelation for reference. (Inset: absorption at B = 10T and laser pulse power spectrum, showing excitation conditions.)